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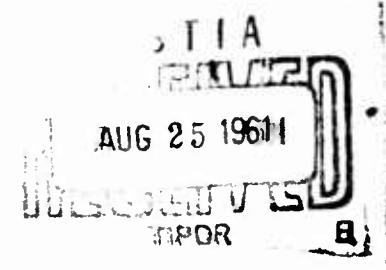
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THE DYNAMIC HYSTERESIS CURVE
OF POLARIZED BARIUM TITANATE ELEMENTS

R. H. Wittekindt



25 July 1961



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LIST OF SYMBOLS (in the order of their appearance)

E = electric field (v/cm, or kv/cm)

E_c = coercive force (kv/cm)

U = voltage (v, kv)

D = charge density on the electrodes of an element (coul/cm², or μ coul/cm²)

F = area of an electrode (cm²)

d = thickness of an element (cm)

P_c = polarization (μ coul/cm²)

Q = charge on the electrodes (μ coul)

ΔQ = incremental charge on the electrodes (μ coul)

Q₀ = P₀ F = initial charge (μ coul)

Q_U = charge on the electrodes of an element with U volts applied in the depolarizing direction (μ coul)

C = capacitance (coul/v = f)

ϵ = dielectric constant (coul/cm v, or μ coul/cm kv)

ϵ_1 = mean value of the dielectric constant along the hysteresis curve

$\epsilon_c = 0.886 \cdot 10^{-13}$ coul/v = dielectric constant of vacuum

α, β = coefficients of the quadratic fit, equation (4)

t = time (μ sec, or nsec)

R = resistance (ohms)

x = capacitance (f), either of a polarized element or of a capacitor for calibration (fig 3)

A, B = voltages (v), as measured in the experimental arrangement (fig 3, sec 5 and 6)

ABSTRACT

The dynamic hysteresis curve of polarized barium titanate elements has been measured in its second quadrant. This is the part of the hysteresis curve $D = D(E)$ which goes from $E = 0$, $D = P_0$ to $E = -E_c$, $D = 0$, whereby the negative field $-E$ increases from zero to E_c within fractions of a microsecond. This part of the hysteresis curve is needed for calculations in connection with shock compression experiments on the barium titanate elements.

1. INTRODUCTION

Extensive investigations have been made on the behavior of polarized barium titanate elements under shock compression. These involve both experiments (ref 1 and 2) and calculations (ref 1, 2, and 3). In order to find certain values (the dielectric constant, for instance) of the compressed material, it is necessary to know the values for the uncompressed, polarized material. This report covers experimental investigations of the hysteresis curve in polarized, uncompressed barium titanate elements.

In a shock experiment, a negative electric field $-E$ (increasing with time) is imposed upon the polarized element in its uncompressed region (that is, before the shock front), in case both electrodes of the element are electrically connected. Simultaneously, the charge density D decreases from its initial value P_0 and reaches $D = 0$ for $E = -E_c$. At this moment, the shock front has traveled all the way through the element and the experiment is over. The process is described in more detail in reference 3. An important feature of this process is that it takes place in fractions of a microsecond.

2. ELEMENTS

The elements were made of barium titanate ceramic, containing 4 percent lead titanate. They are discs 1.6 cm in diameter (area $F = 2.0 \text{ cm}^2$) and 0.33 cm thickness (d). Both flat surfaces are metallized, forming electrodes. The polarization (normal to the electrodes) is applied by the manufacturer, by imposing a high electric field under elevated temperature for a certain time. Barium titanate contains polarized domains, usually distributed in all directions. These domains become aligned under the polarizing treatment and stay aligned afterwards, provided the temperature remains below the Curie temperature (120°C). The whole element is then said to be polarized.

The amount of polarization (P_0) is measured by the amount of charge (Q_0) bound to the electrodes by the influence of this polarization: $P_0 = Q_0/F$. In the present case the mean value of $P_0 = 7.82 \mu\text{coul}/\text{cm}^2$, found by discharging a number of elements at a temperature above 120°C .

The capacitance (C) of the elements is 673 pf if polarized, and 438 pf if not polarized (these are mean values for a number of elements). The dielectric constant, according to the formula $C = \epsilon F/d$ is

$$\epsilon = 1.11 \cdot 10^{-10} \text{ coul/cm} v = 1250 \epsilon_0, \text{ or}$$

$$\epsilon = 0.72 \cdot 10^{-10} \text{ coul/cm} v = 815 \epsilon_0, \text{ respectively}$$

These values were found using a capacitance bridge under very low voltage. Therefore the values of ϵ are limiting values for low field only. Due to these very high values of ϵ , nearly all electric field lines are confined inside the element. This allows neglecting edge effects, and using the most simple formulas:

$$\text{capacitance } C = \epsilon F/d \quad (1)$$

$$\text{electric field } E = U/d \quad (2)$$

$$\text{charge density } D = Q/F \quad (3)$$

with U = voltage across the element (between the electrodes)

Q = charge on the electrodes

3. THE STATIC HYSTERESIS CURVE

The hysteresis curve (in its second quadrant) has been measured statically. Elements were put under a constant negative electric field $-E$ for about 1 min, afterwards discharged (by heat treatment), and the charge measured yielding D . The result is shown in figure 1. The measured points can hardly be connected by a straight line. A quadratic fit is more suitable.

$$D = P_0 + \alpha E - \beta E^2 \quad (4)$$

$$\text{with } P_0 = 7.82 \mu\text{coul/cm}^2$$

$$\alpha = 0.11 \mu\text{coul/cm kv}$$

$$\alpha = \epsilon \text{ from formula (1)}$$

$$\beta = 0.023 \mu\text{coul/kv}^2$$

The linear approximation gives roughly

$$D = P_0 + \epsilon_i E \quad (5)$$

$$\text{with } \epsilon_i = 0.39 \mu\text{coul/cm kv} = 4400 \cdot \epsilon_0$$

Similar values of P_0 , α , β , ϵ_i are found in reference 3 in cases where examples were calculated. From the beginning, however, it was felt that the static curve, according to eq (4) or (5), might not represent the true

\oplus = MEASURED POINTS
 a = QUADRATIC FIT ACCORDING TO EQUATION 4
 b = LINEAR FIT ACCORDING TO EQUATION 5
 c = LIMITING FORM FOR LOW FIELD

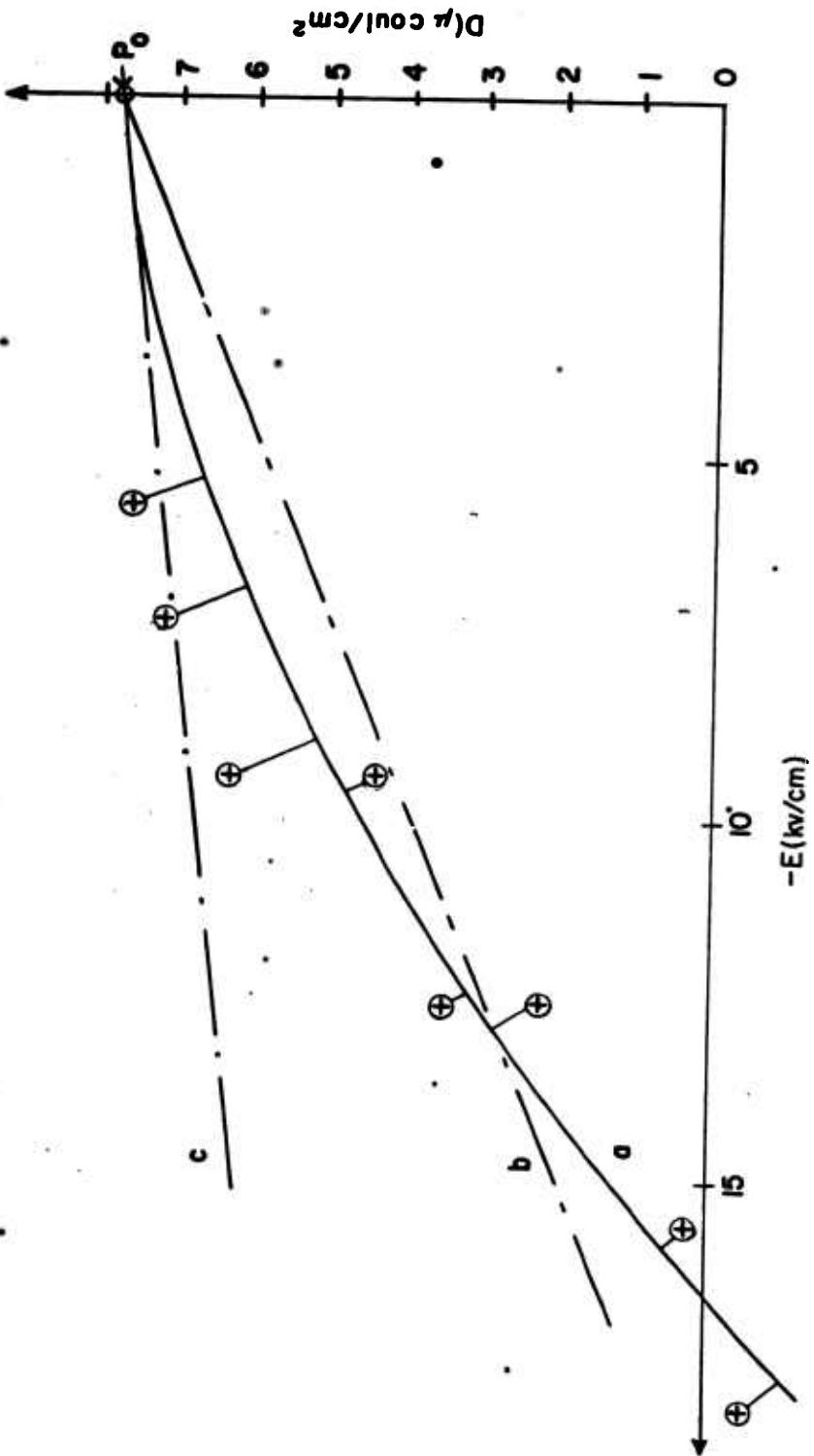


Figure 1. Static hysteresis curve of polarized barium titanate elements (second quadrant)

hysteresis curve for the case encountered in a shock experiment where the field $-E$ rises in fractions of a microsecond to its full value E_c , and where a delay in domain-switching might deform the hysteresis curve.

4. METHOD OF MEASUREMENT

Experiments have been set up to simulate the shock conditions. Elements are exposed to a negative electric field, which increases with time as in a shock experiment. The field ($-E$) and the charge density (D) on the electrodes of the element both are measured as functions of time.

To avoid confusion, the following terms are used throughout: the word "shock experiment" is applied to the case in which a shock front travels through the element. References 1, 2, and 3 deal with shock experiments. The words "hysteresis experiment" or "experiment" are used to describe the arrangement and the measurements in which a rapidly rising negative electric field is imposed upon an element as a whole, thereby simulating the shock conditions in a certain respect. Formulas for calculating the rise of the field $-E$ with time t (in a shock experiment) are given in ref 3 (formulas 3, 9, 33). Unfortunately, these formulas themselves employ the values of the linear or quadratic fit to the proper hysteresis curve i.e., the very values that are to be calculated afterwards from the results of the experiments. To overcome this difficulty, an iteration process can be used. At first the field rise $E_1(t)$ is calculated by the available information, that is, the static hysteresis curve, formula (4) of this report. An experiment is made with a field rise $E_1(t)$. The result is an improved hysteresis curve, better than the static curve, from which an improved function $E_2(t)$ can be calculated. The next experiment is set up to have a field rise $E_2(t)$; the result of this experiment is an even more improved hysteresis curve. If these curves converge to a finite limiting form, the latter is the desired result. In the present case, the curves converge quickly, practically in one step.

Figure 2 shows all the pertinent curves. Curve 1 is the calculated field rise $E_1(t)$ in a shock experiment, if the quadratic fit (formula 4) is the correct one. Actually, it is not, and $E_1(t)$ is the first step only in the iteration process. Curve 2 is the final form of the field rise, the form to which the iteration steps converge. It is calculated using the hysteresis curve as found by the experiments described later. Curves 3 and 4 are the field rise functions as used in the hysteresis experiments. They should fall close to curve 2. Actually, curve 3 is similar to curve 2 in its lower part, curve 4 is somewhat steeper. The resulting hysteresis curve, however, does not depend strongly on the form of the field rise $E(t)$; the hysteresis curves, measured by using a field rise following curve 3 or 4, do not differ from each other. Because curve 3 partly coincides with the correct curve 2, the statement can be made that the hysteresis curve measured along curves 3 and 4 is the same as would be along curve 2. In other words, the hysteresis curve as measured by use of a field rise along curve 3 or 4 is the one that applies to shock experiments.

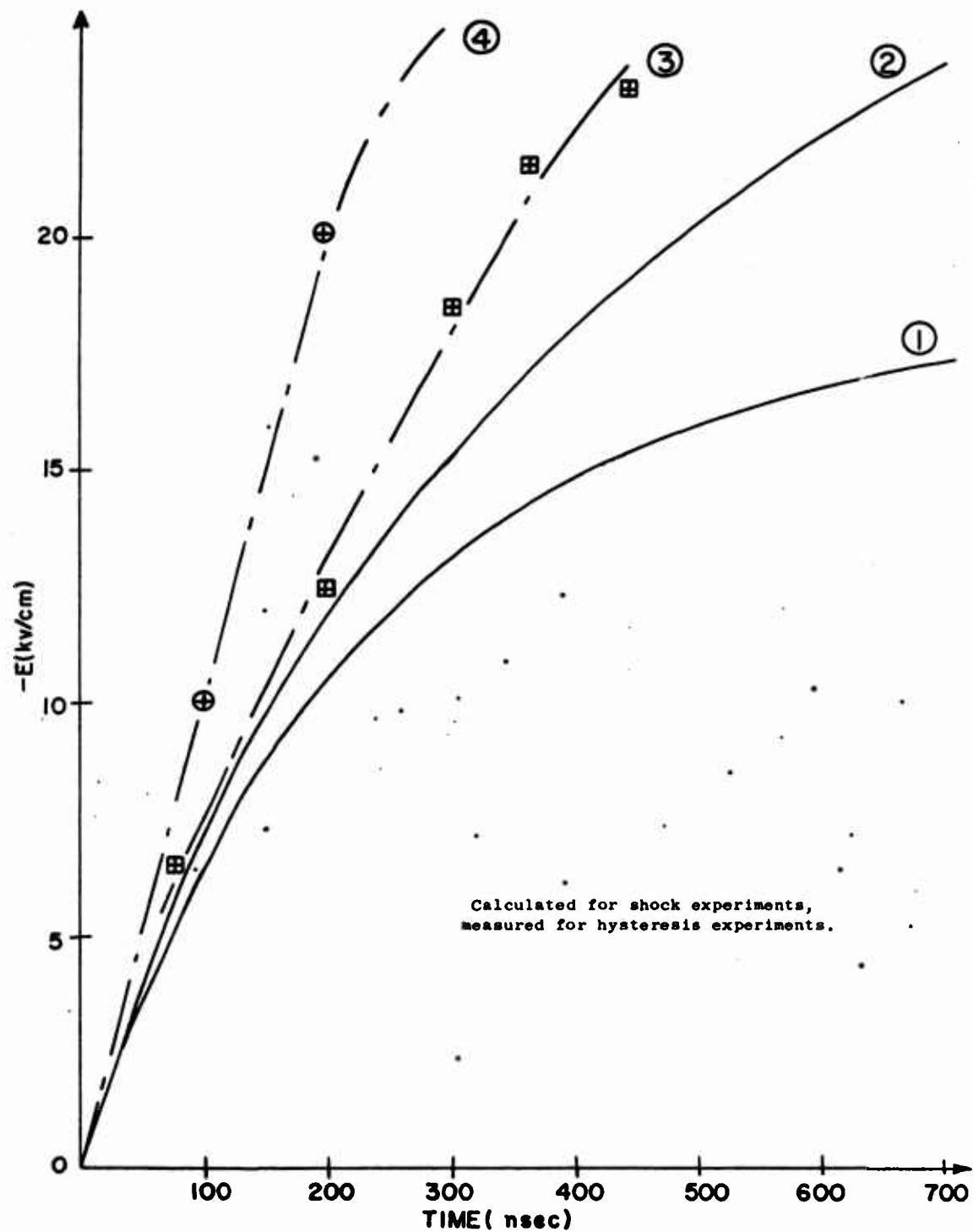


Figure 2. Rise of electric field in elements.

5. EXPERIMENTAL ARRANGEMENT

The experimental arrangement is shown in figure 3. The combination of C_1 , C_2 , C_3 , C_4 and x , as shown results in a capacitance $C = 17.5 \cdot 10^{-9} f$.^{*} The storage capacitor C_0 is charged to U_0 before the experiment starts. By closing the switch S (resulting in a "shot"), the charge on C_0 is distributed over C_0 , R , C , giving a voltage U_1 (across C) which rises from zero to U_0 . Due to the large ratio $C/C_0 = 300$, the value U_1 is nearly U_0 (within 1/3 %). The difference will be neglected so that C is charged up exponentially, or

$$U_1 = U_0 (1 - e^{-t/RC}) \quad (6)$$

with $RC = 0.25 \mu\text{sec}$

The voltages across all the other capacitors rise similarly, with the same time constant. This is especially true for the voltage across x and for the field in x (if an element is in place of x).

$$E(t) = E_{\max} (1 - e^{-t/0.25}) \quad (7)$$

The time constant of curve 2 in figure 2 (if we fit it with an exponential function) is also about 0.25 μsec ; that is, we expect to obtain the desired $E(t)$.

However, in actually trying out this setup, a difficulty arose. In equation (6) the inductance of the loop C_0 , S , R , C is not taken into account. This inductance is larger than that anticipated, a damped oscillation thereby is superimposed on the curve of eq (6). This means for the initial part of the curve a steeper rise and a big overshoot. The cure is to lower U_0 and use the resulting curve as it is. Before, it had been calculated that U_0 must be 6 kv. Now, 2.4 kv (curve 3 in figure 2) and 4.0 kv (curve 4) are used. As has been explained in section 4, the deviation of curves 3 and 4 from 2 (in figure 2) is of no importance to the results. Also, it appears that the voltage (or field strength) rises to a higher final value, but again this is of no importance to the results, because as soon as the field exceeds E_c , the experiment is over.

The rest of the arrangement is explained as follows: C_2 is much bigger than x , therefore (roughly speaking) U_1 measures voltage and field in x , and B (that is the voltage across C_2) measures charge and charge density D on x . The available cathode follower probes are not able to measure a voltage as high as U_1 therefore a voltage-dividing

* C includes x , for which the value $2.14 \cdot 10^{-10} f$ is used. If an element is in place of x , a certain variation of x appears during a shot and results in a much smaller variation in C ; this has been neglected here. The actual measurements, however, are not affected because the voltage across x is measured, not calculated.

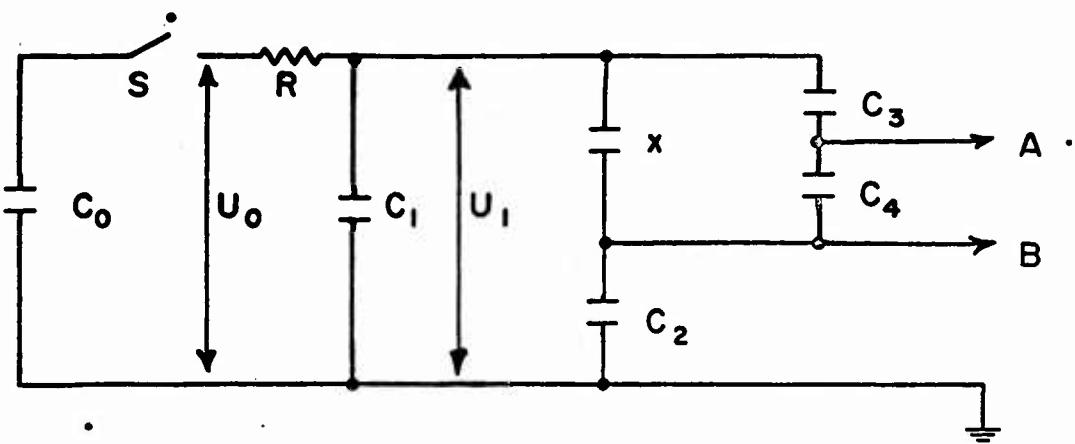


Figure 3. Circuit diagram.

S = switch

U_0 = constant storage voltage across C_0

U_1 = $U_1(t)$ = voltage across C_1

R = 14.54 ohms

$C_0 = 5 \cdot 10^{-6} f$

$C_1 = 15.45 \cdot 10^{-9} f$

$C_2 = 102.35 \cdot 10^{-9} f$

$C_3 = 4.93 \cdot 10^{-12} f$

$C_4 = 50.75 \cdot 10^{-12} f$

X = either a polarized barium titanate element,
or (for calibration) a capacitance of $2.14 \cdot 10^{-9} f$

system C_3 , C_4 is used. The voltage A then stands for the voltage J_1 . The exact formulas are given in section 6.

The outputs A and B are displayed, one at a time, on a Tektronix scope Type 517, using a sweep of 50 nsec/cm. A dual-beam scope was not used because its fastest sweep is only 100 nsec/cm. A Tektronix cathode follower probe P 170 CF is used, in combination with the oscilloscope to measure voltages A and B. All the capacitors (except C_0) are measured on a capacitance bridge. The voltage U_0 is measured by a static voltmeter. The sweep of the scope is calibrated by displaying on the scope a 50-Mc sinewave from a Tektronix Type 180 A Time Mark Generator. The element in position x is covered by oil, to prevent electric breakdown around the edges.

6. DEDUCTION OF FORMULAS

The results of the experimental measurements are the voltages A and B as functions of time. What is wanted however is the field $-E$ inside the element and the charge density D on its electrodes, both as functions of time. Simple formulas can be deduced, to give

$$-E = \text{function of } A, B$$

$$D = \text{function of } A, B$$

The combination of x, C_2, C_3, C_4 forms a capacitance

$$C_6 = \frac{(x + C_5)C_2}{x + C_5 + C_2}$$

the combination x, C_3, C_4 forms a capacitance

$$C_7 = x + C_5$$

where

$$C_5 = \frac{C_3 C_4}{C_3 + C_4}$$

The voltage B is the fraction $C_7/(C_2 + C_7)$ of U_1

$$B = \frac{x + C_5}{x + C_5 + C_2} U_1 \quad (8)$$

The voltage A is the sum of B and the fraction $C_3/(C_3 + C_4)$ of $(U_1 - B)$, which gives

$$A = \frac{x(C_3 + C_4) + C_2 C_3 + C_3 C_4}{x(C_3 + C_4) + C_2 C_3 + C_3 C_4 + C_4 C_2} U_1 \quad (9)$$

Equations (8) and (9) can be solved for x and U_1 yielding

$$x = \frac{C_2 C_3}{C_3 + C_4} \frac{B}{A - B} - \frac{C_3 C_4}{C_3 + C_4} \quad (10)$$

$$U_1 = \frac{C_3 + C_4}{C_3} A - \frac{C_4}{C_3} B \quad (11)$$

The voltage, U , across x is

$$U = U_1 - B$$

$$U = \frac{C_3 + C_4}{C_3} (A - B) \quad (12)$$

The incremental charge ΔQ on the electrodes of x caused by U is

$$\Delta Q = x \cdot U$$

$$\Delta Q = (C_2 + C_4) B - C_4 A \quad (13)$$

The definition of the capacitance of any device is

$$C = \Delta Q / \Delta U$$

the quotient of the incremental charge on the electrodes and the incremental voltage (i.e. U) between these electrodes. For calibration purposes a capacitor $C = x = 2.14 \cdot 10^{-9} f$ is used. For the polarized element the capacity is

$$C = x = \frac{\Delta Q}{U} = \frac{Q_o - Q_u}{U}$$

where

$$Q_o = P_o F$$

Accordingly, equation (13) for a polarized element must be written

$$Q_u = Q_o - \Delta Q$$

$$Q_u = Q_o - (C_2 + C_4) B + C_4 A \quad (13a)$$

*From equations (2) and (12)

$$-E = \frac{C_3 + C_4}{C_3 d} (A - B)$$

and from equations (3) and (13a)

$$D = P_o - \frac{C_2 + C_4}{F} B + \frac{C_4}{F} A$$

Using the values given earlier, and if A and B are expressed in volts

$$-E = 0.03415 (A - B) \quad \text{in kv/cm} \quad (14)$$

$$D = 7.82 - 0.051 B \quad \text{in } \mu\text{coul}/\text{cm}^2 \quad (15)$$

The term, $\frac{C_4}{F} A$, in equation (15) is dropped since $C_2 > 2000 C_U$.

The voltages A and B are found as vertical deflections on the screen of the scope. The calibration between the geometrical deflection and the voltage is done in the following way. If a capacitor of known value is put in place of x, everything necessary can be calculated. In this case equations (8) and (9) are used, with $x = 2.14 \cdot 10^{-9} f$. This gives (in volts)

$$A = 0.107 \cdot 25 \cdot U_1$$

$$B = 0.020 \cdot 52 \cdot U_1$$

The particular value of $U_1 = U_o$ that is determined by an electrostatic voltmeter is used in the calibration procedure. The sweep time of the scope is changed to $2\mu\text{sec}/\text{cm}$. Thus the rise of the curve $U_1(t)$ and the oscillations due to the inductance are over after a sweep of 2 cm, and the rest of the sweep is a straight line of constant height, representing $U_1(t) = U_o = \text{constant}$. Then we have

$$A = 107.25 \cdot U_o \quad (16)$$

$$B = 20.52 \cdot U_o$$

with A, B in volts, if U_o is read in kilovolts.

7. EXPERIMENTAL RESULTS

Four sets of experiments have been performed, in which 22 elements were tested. After each shot the element has to be replaced by a new one.

* The shot partially depolarizes the element. The final polarization is $5.5 \mu\text{coul}/\text{cm}^2$ compared with an initial value of $7.8 \mu\text{coul}/\text{cm}^2$.

Sets I and IV were taken with $U = 4$ kv (along curve 4 in figure 2), sets II and III under $U = 2.4$ kv (along curve 3). In 10 of these shots voltage A was displayed on the scope, in the other 12 shots voltage B was displayed. For each shot a photograph of the trace on the screen was taken, which afterwards was transferred onto graph paper, taking into account the sweep-time-calibration, so that the trace on the paper was linear with time (x-deflection).

The vertical distance of the trace from the base line (y-deflection) was then measured on the paper at intervals of 25 nsec (which amounted to steps of about 0.5 cm along the original trace on the screen). This vertical distance y was transformed into voltage A or B by use of the calibration (equations 16 in section 6), which had been made before and after each set of shots. Corresponding points of the curves A in one set of shots were averaged. The same was done with curves B. This left one curve A and one curve B for each set, in steps of 25 nsec. Now the formulas (14) and (15) were employed, which gave $-E$ and D both as functions of time in steps. For any one of these time steps the proper values $-E$ and D were plotted as points in the E-D diagram, giving the hysteresis curve in its second quadrant. By repeating this process for all of the four sets of shots, there resulted four curves $D = D(E)$, which were nearly identical and could be represented by one curve. Figure 4 shows all the plotted points and the resulting hysteresis curve.

To describe this curve by a numerical formula, it is found that neither a linear nor a quadratic fit is fully suitable. The following two expressions do fit the curve b in figure 4. Between $E = 0$ and $E = -5.5$ the hysteresis curve follows a quadratic expression

$$D = 7.82 + 0.11 \cdot E - 0.023 \cdot E^2 \quad (17)$$

Between $E = -5.5$ and $E = -E_c = -23.6$ kv/cm (and beyond that point) the hysteresis curve follows a linear expression

$$D = 8.50 + 0.36 \cdot E \quad (18)$$

The two expressions meet tangentially at $E = -5.5$ kv/cm, $D = 6.52 \mu\text{coul}/\text{cm}^2$, $t = 50$ nsec. It may be noted that the static hysteresis curve (eq 4) follows the same expression (17), but up to the maximum field $E = -E_c = 16.6$ kv/cm. The dynamic hysteresis curve, on the other hand, is as a whole better described by the linear fit, equation (18).

8. CONCLUSIONS

The results of the experiments show that the hysteresis curve of polarized barium titanate elements is changed if the electric field $-E$ rises very fast. For a slow change of $-E$, the hysteresis curve in the second

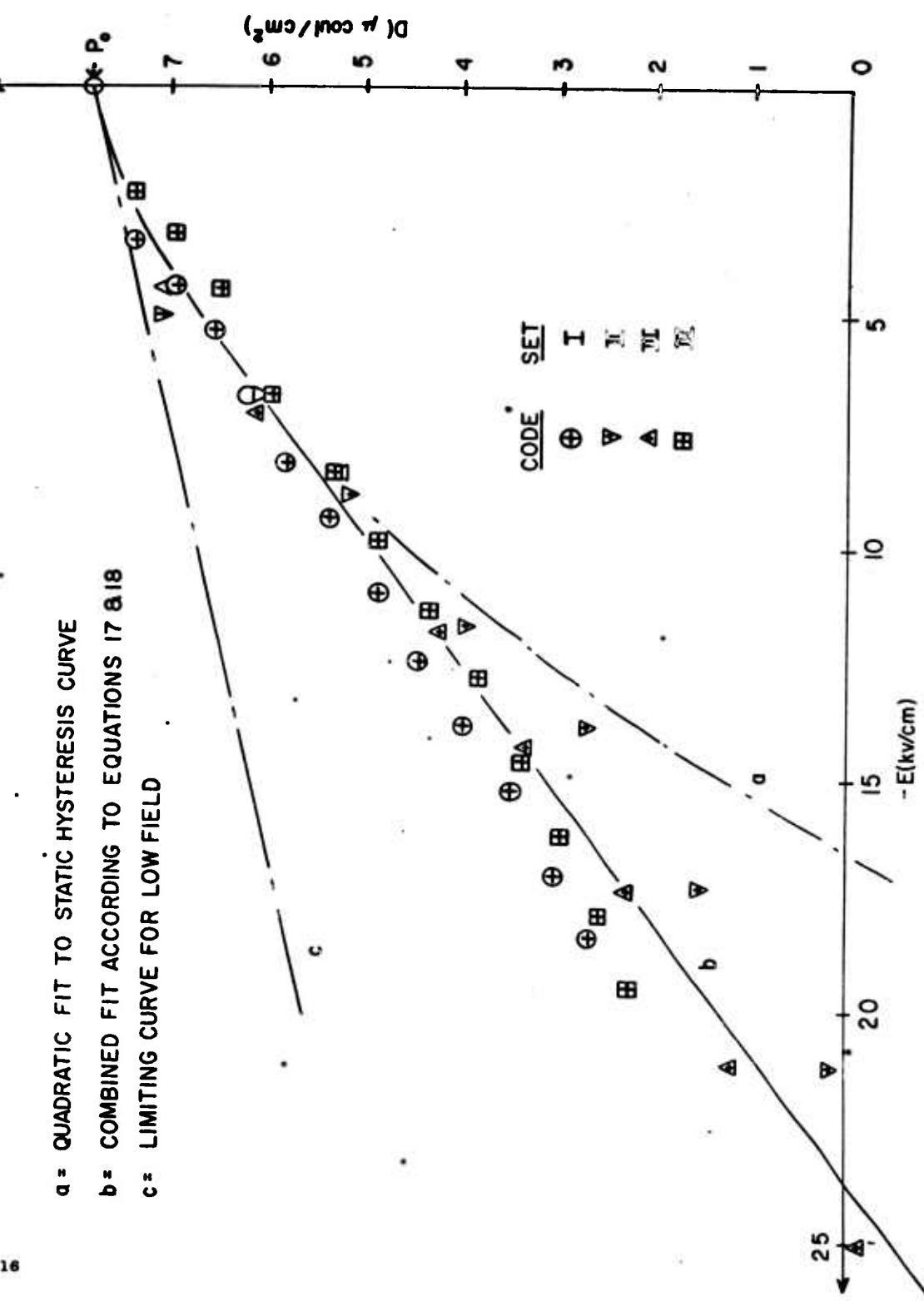


Figure 4. Dynamic hysteresis curve of polarized barium titanate elements (second quadrant).

quadrant can be described by a quadratic fit. If $-E$ rises approximately exponentially with a time constant of 0.25 μsec , then the hysteresis curve follows the quadratic formula only up to 0.05 μsec , and follows a linear formula from there on. This means that a higher field $-E$ is needed to discharge the element (to bring D to zero) in this high-speed experiment, than is needed in a slow experiment. This may be explained by a delay in domain switching.

As far as shock experiments are concerned, results show that the linear fit is best. Only the very first part of the output curve of a shock experiment then must not be taken into account, up to 0.05 μsec . The linear fit facilitates the evaluation of the results from shock experiments considerably.

The value of the dielectric constant of polarized barium titanate elements under fast-rising high-negative electric field is

$$\epsilon_i = 0.36 \mu\text{coul}/\text{cm kv}^-$$

$$\epsilon_i = 3.6 \cdot 10^{-10} \text{ coul}/\text{cm v}$$

$$\epsilon_i = 4060 \cdot \epsilon_0$$

(ϵ_0 = dielectric constant of vacuum).

If the linear formula is used to evaluate shock experiments, then the apparent value of the polarization (for the purpose of such an evaluation only)

$$\begin{aligned}\overline{P}_0 &= 8.50 \mu\text{coul}/\text{cm}^2 \\ &= 8.50 \cdot 10^{-6} \text{ coul}/\text{cm}^2\end{aligned}$$

has to be used, rather than the actual initial polarization

$$P_0^* = 7.82 \mu\text{coul}/\text{cm}^2$$

ACKNOWLEDGMENTS

The author wishes to thank Mr. P. S. Brody for suggestions and discussions in connection with the experiments described in this report. The work was supported by funds of the Transducer Project.

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Titanates Barium compounds

The dynamic hysteresis curve of polarized barium titanate elements has been measured in its second quadrant. This is the part of the hysteresis curve $D = D(E)$ which goes from $E = 0$, $D = P_0$ to $E = -E_c$, $D = 0$, whereby the negative field $-E$ increases from zero to E_c within fractions of a microsecond. This part of the hysteresis curve is needed for calculations in connection with shock compression experiments on the barium titanate elements.

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